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IASI measurements of reactive trace species in biomass burning plumes

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Abstract. This work presents observations of a series of short-lived species in biomass burning plumes from the Infrared Atmospheric Sounding Interferometer (IASI), launched onboard the MetOp-A platform in October 2006. The strong fires that have occurred in the Mediterranean Basin – and particularly Greece – in August 2007, and those in Southern Siberia and Eastern Mongolia in the early spring of 2008 are selected to support the analyses. We show that the IASI infrared spectra in these fire plumes contain distinctive signatures of ammonia (NH_3), ethene (C_2H_4), methanol (CH_3OH) and formic acid (HCOOH) in the atmospheric window between 800 and 1200 cm^{-1} , with some noticeable differences between the plumes. Peroxyacetyl nitrate ($\text{CH}_3\text{COOONO}_2$, abbreviated as PAN) was also observed with good confidence in some plumes and a tentative assignment of a broadband absorption spectral feature to acetic acid (CH_3COOH) is made. For several of these species these are the first reported measurements made from space in nadir geometry. The IASI measurements are analyzed for plume height and concentration distributions of NH_3 , C_2H_4 and CH_3OH . The Greek fires are studied in greater detail for the days associated with the largest emissions. In addition to providing information on the spatial extent of the plume, the IASI retrievals allow an estimate of the total mass emissions for NH_3 , C_2H_4 and CH_3OH . Enhancement ratios are calculated for the latter relative to carbon monoxide (CO), giving insight in the chemical processes occurring during the transport, the first day after the emission.

1 Introduction

Most of the processes driving atmospheric chemistry occur on time scales of seconds to days. The species involved, highly reactive, are present in the atmosphere in small concentrations and are mostly hard to track in space and time. When released at the surface as primary pollutants, they react rapidly close to the source region, except for a small fraction that survives boundary layer chemistry, escapes to the free troposphere and is transported to regions downwind. The distance travelled strongly depends upon the species chemical lifetime, the latter being in turn strongly influenced by the local chemical and thermodynamic conditions.

Of particular interest is the monitoring close to the surface of constituents which impact, directly or not, human health and ecosystems. These species are controlled in industrialized countries by air quality standards, which are defined in terms of the ambient abundance of ozone (O_3), nitrogen oxides (NO_x), sulfur dioxide (SO_2), carbon monoxide (CO) and particulate matter ($\text{PM}_{2.5}$ and PM_{10}). Ozone itself is a secondary pollutant which is formed by complex reaction mechanisms involving NO_x , methane (CH_4), CO and a large series of volatile organic compounds (VOCs).

Ground-based instruments located nearby source regions, such as urban areas, are obviously best suited for measuring reactive species in the boundary layer, as they offer at the same time a high sensitivity (down to a few parts per trillion – 10^{-12} – in fractional concentration), accuracy and repetitiveness. They are therefore important for the local air quality surveillance and forecast systems. However, they are unable to resolve and track pollution plumes in space and time and can only partly contribute, for instance, to the study of transboundary transport of pollution. In recent years,



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nadir-looking satellite instruments have greatly helped drawing the grand picture of tropospheric pollution by measuring a series of primary and secondary reactive gaseous pollutants down to the surface, from the local to the global scale (Martin, 2008 and references therein). Routine observations include NO_2 , SO_2 , H_2CO , $\text{C}_2\text{H}_2\text{O}_2$ and halogen compounds from UV-visible sounders (Wagner et al., 2008 and references therein), and principally O_3 , CO and CH_4 from infrared sounders (e.g. Clerbaux et al., 2003, 2009). Recently the possibility of probing NH_3 and methanol (CH_3OH) from TIR radiances was demonstrated (Beer et al., 2008), adding to the potential of TIR sounding missions in contributing to monitor the fast chemistry of the lower troposphere. Considering this, the recently launched Infrared Atmospheric Sounding Interferometer (IASI) onboard MetOp-A, which has unique spatial resolution and sampling, is expected to benefit the surveillance of the atmospheric system, operationally, and from a local to a global scale (Clerbaux et al., 2007).

In this work, we intend to go a step further by showing that TIR sounders can be used to identify and quantify a range of short-lived chemical species that are not observed in background atmospheric situations. For this purpose, we focus our study on IASI measurements above fire plumes from the 2007 wildfires in the Mediterranean basin (Boschetti et al., 2008; Turquety et al., 2009) and the spring 2008 boreal fires in Southern Siberia – Baikal Lake area (Warneke et al., 2009) – and Eastern Mongolia. Fire plumes are chosen because of the expected elevated concentrations of gases and particles released simultaneously in the boundary layer and because of the usual high reactivity of these extreme environments (e.g. Andreae and Merlet, 2001). Large biomass fires are furthermore known to significantly affect air quality conditions prevailing locally, especially when the plume is confined in the boundary layer, but also regionally when the plume is emitted in the free troposphere and transported downwind over long distances (Langmann et al., 2009 and references therein). The local impact is mostly due to the release of particles which remain close to the emission point, whereas the regional impact is mostly related to the emission and transport of ozone precursors, such as CO , NO_2 , and VOCs, the chemistry of which can induce large surface ozone concentrations farther away (Langmann et al., 2009; Pfister et al., 2008; Morris et al., 2006).

The next section provides an overview of the measurements and the relevant theoretical elements. These include a brief description of IASI, of the two fire events examined here, and of the inverse method used to retrieve trace gas abundances from the spectra. Section 3 reports on the detailed spectroscopic analysis of different fire plumes, focusing on the assignment of spectral features to species rarely measured before from satellite observations. This section includes a focused discussion of the Greek fires, giving insights on the chemistry processes occurring during the first 24 h after the plume's emission. We then draw conclusions

and open perspectives for further research, notably in regard to the possibility to obtain global distributions for some of the newly species observed.

2 Measurements

2.1 IASI

A detailed description of IASI is provided by Clerbaux et al. (2009) in this special issue. As compared to other thermal infrared sounders in orbit, the IASI advanced Fourier transform spectrometer, offers a large and continuous spectral coverage of the infrared region ($645\text{--}2760\text{ cm}^{-1}$), at a medium spectral resolution (0.5 cm^{-1} apodized). The IASI spectra are dominated by rotation-vibration transition lines of H_2O , CO_2 , CH_4 , N_2O , O_3 and CO , and also allow providing key information on surface properties using atmospheric windows. Given the high radiometric performances, weak absorption features such as for instance HNO_3 (Wespes et al., 2009) and CFCs (Coheur et al., 2003) are seen on each individual observations. The spectral information contained in the measurements are used to tackle the primary objective of IASI, which is to support operational numerical weather predictions by providing accurate and highly resolved vertical profiles of temperature (from the CO_2 lines) and humidity (from the H_2O lines) (Schlüssel et al., 2005). The observations of greenhouse gases and reactive species in the IASI spectra, and in particular O_3 and CO allow climate and atmospheric chemistry issues to be tackled as well, also opening perspectives for operational applications (Clerbaux et al., 2007).

The IASI sounder currently in orbit is the first of a series of three successive instruments that are part of Europe's polar orbiting meteorological satellites (MetOp), dedicated for operations in the 2006–2020 timeframe. Being designed primarily to meet the needs of meteorology, it provides a global coverage of the Earth surface twice daily with a relatively small pixel size on the ground (12 km at nadir). This high spatial and temporal sampling capability along with the spectral and radiometric performances of the FTS, offer a unique support for identifying local and sudden emissions at the surface and for following the fate and transport of the resulting pollution plumes. Typical applications are for instance found in the monitoring of volcanic eruptions (Clarisse et al., 2008) and fires (Turquety et al., 2009), using signatures of SO_2 and CO , respectively.

2.2 Radiance and brightness temperature spectra

Thermal infrared nadir sounders measure the radiation emitted by the Earth, modified by atmospheric extinctions (absorption and scattering) and emissions along the path. The spectra measured at the top of the atmosphere are expressed as radiance ($\text{W/cm}^2\text{ sr cm}^{-1}$), which represent the outgoing flux collected within a solid angle at a given wavenumber,

per unit of surface. The radiance spectra can be conveniently transformed to brightness temperature spectra, by inverting the source function, usually represented by a grey body (Planck blackbody function multiplied by the surface emissivity). In a so-called window channel without atmospheric absorption, the brightness temperature associated to a scene with unitary emissivity would be equal to the Earth's surface temperature. Extinction and emission processes occurring in the atmosphere at a given wavenumber $\tilde{\nu}$ cause the brightness temperature to be respectively lower and higher than the surface temperature in a nearby window channel. Accordingly, the difference in brightness temperature between a perturbed and a reference window channel, hereafter BTd, can be used as a probe for the presence of absorption or emission features in the spectra. The use of BTds is common for multispectral sounding and represents a fast and robust method for the identification and tracking of sudden events such as volcanic plumes (Clarisse et al., 2008).

2.3 Concentration measurements

Retrieving trace gas abundances from IASI spectra is done by adjusting onto the measurement a simulated radiance spectrum, computed from the radiative transfer equations using our best knowledge of the atmospheric state at the time and place of the observation. In the forward model, a particular care has to be given to the local meteorological conditions, characterized by pressure, temperature and humidity profiles, as well as to the surface type and emissivity. For this work we use pressure, temperature and humidity vertical profile (Level 2) products disseminated operationally by EUMETSAT (Schlüssel et al., 2005). The preliminary validation of these level2 meteorological products reports an accuracy close to the mission objectives: for temperature the error is 0.6 K in the free troposphere, increasing to 1.5–2 K at the surface and in the upper troposphere with a bias of ± 0.5 K, while for the relative humidity the error is 10% with a bias within $\pm 10\%$ (Pougatchev et al., 2009). These parameters are kept fixed in the inversion process, except for the surface temperature and the humidity profiles which are adjusted to provide the best possible fits in the spectral regions of interest. For the Greek fires in 2007, the Level 2 data were not available and the pressure and temperature data for the corresponding scenes were therefore taken from the European Center for Medium Weather Forecast (ECMWF), interpolated to match the time and place of the measurements. For the emissivity, an average value from the 12 channels from MODIS/Terra climatology are used (Wan, 2008). The spectroscopic parameters, including line parameters and absorption cross sections for CFCs, are from the HITRAN database (Rothman et al., 2005). For CH_3COOH , missing in HITRAN, the cross sections from the PNNL-Vapor phase infrared spectral library (Sharpe et al., 2004) are used. Also the water vapour and carbon dioxide are taken into account using the MT-CKD formulations (Clough et al., 2005).

As nadir measurements integrate all photophysical processes occurring over the vertical altitude range of the atmosphere, one can typically hope to extract total column amounts. For a given species, this is usually done by scaling a reference profile iteratively such as to reproduce the amplitude of the observed spectral features as close as possible. For some gases, however, weakly resolved vertical profiles can be derived as well, considering the pressure and temperature dependences of their spectra. The problem becomes unfortunately ill-conditioned and can only be solved by using constrained retrieval approaches. The Optimal Estimation (Rodgers, 2000) is the most widely used method for remote sensing purposes. It is implemented in the *Atmosphit* software, which includes a full line-by-line radiative transfer model, as well as in the FORLI (Fast Operational/Optimal Retrieval on Layers for IASI) dedicated software for near-real time and large scale processing of IASI since 2007. In this work, we have used the *Atmosphit* software for all species except CO, which is routinely processed by FORLI-CO in the group (George et al., 2009). The theoretical approaches of both algorithms are extensively described elsewhere (Clarisse et al., 2008; Coheur et al., 2005; Turquety et al., 2009; Wespes et al., 2009).

The trace gases profiles used for the forward model and the retrievals with *Atmosphit* in the atmospheric window are from the US standard atmospheres (US Government Printing Office, 1976), scaled in the case of CO_2 to a vmr of 385 ppmv. Exceptions are O_3 , CH_4 and HNO_3 , for which the prior are global averaged yearly profiles from climatology, used along with the associated covariance matrices (Turquety et al., 2004; Wespes et al., 2009) and for CH_3OH and C_2H_4 for which we use global averaged profiles from the LMDz model (Hauglustaine et al., 2004). For these two species and for NH_3 , which are central to this paper, we allow a large variability in the retrieval of vertical profiles (6 layers of 3 km thickness are retrieved although the objective is not to deduce vertically resolved information due to the weak signals). This is done by using an ad-hoc covariance matrix with very large variability (diagonal elements) and by considering for the off-diagonal elements a simple exponential decay with 7 km correlation length. These assumptions on the variance-covariance matrix, which are required to capture the large values of concentrations in the plume are unrealistic in the geophysical sense, and do accordingly not allow for a comprehensive posterior characterization of the retrieved products in terms of vertical sensitivity and errors. Hence averaging kernels will not be shown in the following and the reported retrieval errors, which do not properly represent the vertical smoothing and do not account for the possible impact of e.g. aerosols, should be taken with caution. As a result of this also, some of the retrieved quantities which will be discussed next, in particular the total emitted mass, should mainly be considered as indicative.

2.4 Selected fire events

This paper concentrates on two large fire events that occurred in the Northern Hemisphere since the IASI launch in October 2006: the August 2007 wildfires in the Mediterranean basin and the April–May 2008 boreal fires in Southern Siberia and Eastern Mongolia, in the vicinity of the Baikal Lake.

The wildfires in the Mediterranean basin are relatively well documented (Boschetti et al., 2008; EFFIS, 2008) and also discussed more thoroughly by Turquety et al. (2009). They lasted for several days and burned close to 900 thousands hectares, with about one third in Greece (EFFIS, 2008), which experienced that year the worst fire situation on record. In Greece the fires have been extremely severe due to combination of persistent high temperatures (3 consecutive heat waves), drought and strong winds. The fires were particularly intense in the last week of August 2007 in the Peloponnese (22 to 30 August), with five major fires burning 170 000 hectares (ha) and releasing a large quantity of gases and particles in the atmosphere. Unusually elevated CO, well above several parts per million by volume mixing ratios in the boundary layer, have for instance been reported close to the fire source (Turquety et al., 2009). Rapid transport at relatively low altitude (estimated around 3–5 km) caused increased CO level above most part of the Mediterranean basin during these days.

The wildfires in Kazakhstan and Southern Siberia in April 2008 were also exceptional, starting earlier than usual due to relative high temperatures and low levels of precipitation. Before the end of April, more than 900 large-scale fires were reported, with 36 000 ha of forest being devastated, likely resulting from human negligence. This event was amongst the worst that Russia experienced in the last 30 years. Large fires were in particular reported in the area between the Baikal Lake and the Amur River between 17 and 24 April. Unlike the ones in Kazakhstan Southern-Siberia region, which were attributed to agricultural practices, those in the Baikal area are likely due to forest fires (Warneke et al., 2009). Another relatively important burning episode occurred later in mid May in a nearby region in Eastern Mongolia, lasting a few days only. Besides their importance for local air quality, pollutants emitted by these high latitude fires in the Northern Hemisphere are also important due to rapid transport to the remote Arctic (Stohl, 2006), playing a role in the development of the so-called Arctic haze and the lowering of the surface albedo (Law and Stohl, 2007; Generoso et al., 2007). This has been the case for the Baikal fires of April, which have been monitored by the NOAA WP-3D aircraft during the airborne field experiment ARCPAC (Aerosol, Radiation and Cloud Processes affecting Arctic Climate), over Northern Alaska and the Arctic sea ice (Warneke et al., 2009). The transported plumes were sampled by a suite of airborne instruments at relatively low altitude, from the surface to 6.5 km in the free troposphere, which corresponds to the highest flight altitude for the WP-3D (Warneke et al., 2009).

3 Results

3.1 Spectroscopic analyses

Figure 1 presents IASI observations of a fire plume originating from the Baikal region (49.50° N, 110.32° E), on 18 April 2008, around 20:50 local time. Two brightness temperature spectra are highlighted (red and blue curves), covering the atmospheric window from 800 to 1200 cm⁻¹ and the CO 1-0 rotation-vibration band from 2050 to 2220 cm⁻¹. Residual spectra are also shown. These were calculated by subtracting from the two target spectra within the plume a spectrum recorded nearby in background conditions (grey lines in Fig. 1), which was chosen to have similar surface temperatures and ozone and humidity concentrations. This allows the removal of all major background lines in these spectral regions. The residual spectrum in red shows strong features emerging from the background, all with negative BTD values, which indicate the occurrence of additional absorptions in the fire plumes. On the shortwave end these spectral features are rotational lines of the CO fundamental band. Their large negative BTD values of –10 K and below indicate strong concentration enhancements of that species in the fire plume. In the atmospheric window, the residual features are principally due, as will be shown next, to ammonia and several volatile organic compounds, not observed in the background spectra. Interestingly, the spectrum plotted in blue in Fig. 1 shows the same features but with positive BTD values, suggesting the presence of an emitting layer of gases and/or particles in the path, at a temperature higher than that of the source. This would be consistent with the IASI temperature profiles provided by EUMETCast, which shows an inversion layer at about 2.2 km (Fig. 1, right panel), and may give indication, as extensively discussed in Clarisse et al. (2009b), of the altitude of the plume at this location, i.e. within or just above the boundary layer. More generally, the temperature variations in the lowest layer of the troposphere and in particular the difference in temperature between the surface and the air just above it (the thermal contrast) is well known to strongly regulate the sensitivity of infrared sounders in the low troposphere. The spectrum with strong absorption lines in Fig. 1 (shown in red with negative BTD values) has for instance a surface temperature of 296 K and a much lower air temperature of 272 K (right panel of Fig. 1); the resulting high positive thermal contrast greatly enhances the sensitivity of the sounder to the surface. If the emitted fire plume was in the boundary layer, this higher sensitivity would be a good demonstration of IASI probing the lowermost atmospheric layers. The observation of South Siberian fire plumes made later above Alaska and the Arctic Sea below 6.5 km (Warneke et al., 2009) tend to confirm that the plume was emitted and transported at relatively low altitude. It is worth pointing out that the situation described here, where we would have observed for an evening scene both emission and absorption spectra with contribution from the boundary

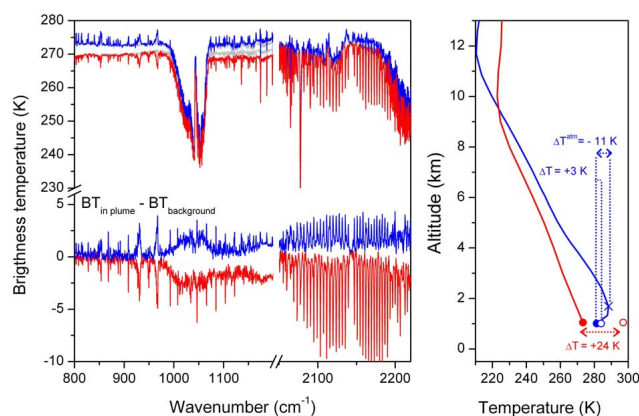


Fig. 1. Left: example of IASI observations of ammonia in a fire plume in the vicinity of the Baikal Lake (see Fig. 2), on 18 April 2008, around 20:50 local time. Two example spectra within the plume are shown, with focus on the spectral regions from 800 to 1200 cm^{-1} and 2050 to 2220 cm^{-1} . The residual spectra in the Figure are obtained by subtracting nearby background spectra, shown in grey line, with similar temperature, humidity and ozone (note the weak presence of the latter in the residual between 1000 and 1100 cm^{-1}). The residuals show both positive (blue) and negative values (red), corresponding to unusual emission and absorption features from the fire plumes. Above 2100 cm^{-1} , these clearly show the enhancements of CO concentrations as compared to background situations. In the atmospheric window, these features are attributable to NH_3 and a series of VOCs. Right: temperature profile corresponding to the two fire spectra, with the thermal contrast (ΔT) values indicated. The temperatures are those disseminated operationally by EUMETCast.

layer is uncommon, as IASI shows in general greater sensitivity to the surface during its morning orbit, when the thermal contrast can reach high positive value (Clarisse et al., 2009b). On 18 April the observations of remarkable emission/absorption features in the IASI spectra extend over a wide area, located between the Baikal Lake and River Amur, as displayed in Fig. 2 using BTDs between a channel sensitive to NH_3 at 867.75 cm^{-1} and two window channels located on each side of it (at 861.25 cm^{-1} and 873.50 cm^{-1}).

Figure 3 shows spectral fits in the region 800–1200 cm^{-1} , for the spectrum plotted in Fig. 1. The fitted spectrum was obtained by adjusting the columns or profiles for all species absorbing in that spectral region (O_3 , H_2O , CO_2 , HNO_3 , CFC11 and CFC12) along with surface temperature, but excluding ammonia (NH_3), ethene (C_2H_4) and methanol (CH_3OH). The assignment of the enhanced features in the plume spectra seen in Fig. 1 to these three compounds becomes unambiguous when comparing the residual spectrum (observed-fitted, middle panels of Fig. 3), to the calculated transmittance of the missing species (bottom panels of Fig. 3). The NH_3 spectrum is particularly visible, extending over almost 400 cm^{-1} , with two noticeable Q -branches and several strong lines, all belonging to the ν_3

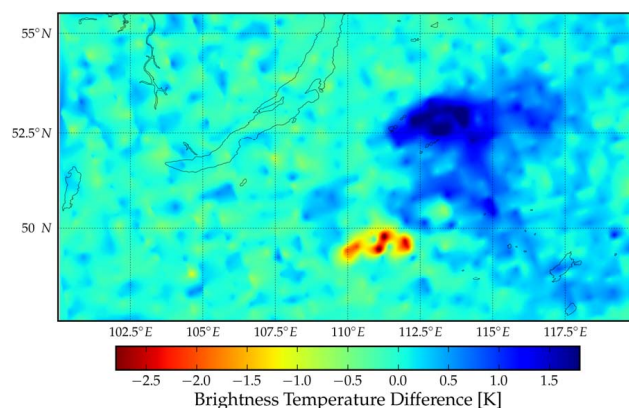


Fig. 2. IASI observations of ammonia in a fire plume in the vicinity of the Baikal Lake, on 18 April 2008 (20:50 local time). The figure shows the spatial distribution of the plume, using the NH_3 signal, expressed in brightness temperature difference between a channel sensitive to NH_3 at 867.75 cm^{-1} and two window channels at 861.25 cm^{-1} and 873.50 cm^{-1} . Note the positive and negative values, representing features in emission and absorption, respectively. The spatial extension of the plume is revealed by the high values, above +0.8 K and below -0.8 K; the values lying in between these thresholds are mostly representative of the noise level. Two typical spectra from these plumes are displayed in Fig. 1.

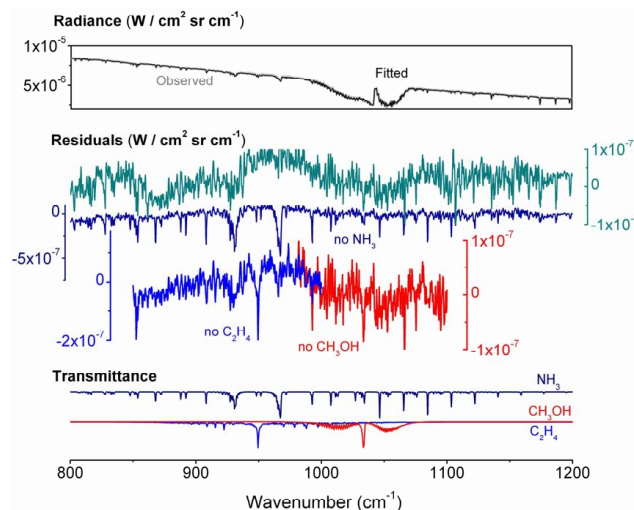


Fig. 3. Top: observed and fitted IASI radiance spectra from the fires in the Baikal area (observation made on 18 April at 49.50° N, 110.32° E; see Fig. 1 red curve), with all species included in the retrieval except NH_3 , C_2H_4 and CH_3OH . Middle: spectral residuals (observed-fitted spectrum) with all species in the retrieval (dark cyan), without NH_3 (dark blue), without C_2H_4 (blue) and without CH_3OH (red). Bottom: simulated transmittance for NH_3 , C_2H_4 and CH_3OH alone, with arbitrary concentrations. See text for details.

band. For C_2H_4 and CH_3OH , it is essentially the Q -branch, of respectively the ν_7 and ν_8 bands, that show up. One should point out that the CH_3OH spectrum is strongly overlapped by that of ozone.

Figure 4 provides a similar comparison in the region from 1070 to 1250 cm^{-1} , which is a relatively transparent window, containing several water lines of medium intensity and additional weaker contribution of O_3 , N_2O and CFC12. In the plume, however, this portion of the spectrum is significantly affected by broadband extinctions. In Fig. 4 this is shown for two spectra, one which is the same as that of Figs. 1 and 3, the other corresponding to a fire plume from 17 May in Eastern Mongolia (46.09° N , 118.99° E , 11:07 local time), also revealing signatures of NH_3 , CH_3OH and C_2H_4 . Among these broadband features, one, which is mostly visible in the 17 May spectrum, is unambiguously attributable to formic acid (HCOOH) in its ν_6 band, with a marked Q -branch at 1105 cm^{-1} . Further extinction, on the shortwave end of this spectral portion, is tentatively assigned here to acetic acid (CH_3COOH), possibly amplified by peroxyacetyl nitrate (PAN), both with absorptions in the CO stretching mode. The assignment to PAN is in fact more certain for the 17 May plume, considering that an additional spectral band of that species (NO_2 bending mode) is well visible between 760 and 830 cm^{-1} (middle vertical panel in Fig. 4). For this spectrally rich plume the following total columns are retrieved: $[\text{NH}_3]=5.7\pm0.1$, $[\text{C}_2\text{H}_4]=2.2\pm0.3$, $[\text{CH}_3\text{OH}]=5.1\pm0.7$, $[\text{HCOOH}]=5.2\pm0.2$, $[\text{CH}_3\text{COOH}]=0.6\pm0.2$, $[\text{PAN}]=2.0\pm0.1$, all expressed in $10^{16}\text{ molecules cm}^{-2}$. We stress that the errors given here on the columns are the statistical (1σ) retrieval errors, which may not be representative considering the assumptions made on the variance-covariance matrices (see Sect. 2.3.) and the fact that possible interferences by e.g. aerosols, have not been taken into account. Furthermore, although vertical profiles are retrieved, the number of independent pieces of information (the so-called degrees of freedom for signal – DOFS – in the Optimal Estimation Method) is for most species not larger than one, suggesting that only a column can be retrieved. Only in the case of NH_3 , for which the signal is strongest, the DOFS gets occasionally to 1.5 but this value remains tributary, however, of the large variability allowed in the retrieval. In the following we therefore only use column abundances. We note in addition that other classic fire tracers, including C_2H_2 , C_2H_6 , H_2CO , HCN and CH_3CN have not been identified in these IASI spectra. For C_2H_2 and HCN this could be due in part because their main absorption bands lie in a CO_2 -saturated region of the spectrum below 750 cm^{-1} , while for H_2CO this is explainable to the weak lines in the near infrared above 2500 cm^{-1} , where the noise performances of IASI are significantly reduced. For C_2H_6 and CH_3CN the reason is unclear and in all cases not attributable to instrumental limitations.

Comparing the different fire plumes, it is worth stressing that unlike the spectra of 18 April in the Baikal region, the

temperature profile for the 17 May case in Eastern Mongolia is not associated with a favourable thermal contrast situation (a ΔT of -6 K prevails for the example spectrum of Fig. 4). The observation of NH_3 and VOCs in these fires may therefore point to a higher altitude of the plume, well above the boundary layer, where the IASI measurements are by nature more sensitive. The injection of fire plumes directly into the free troposphere can take place through different convection mechanisms (e.g. Freitas et al., 2007), but are relatively infrequent (Freitas et al., 2007; Labonne et al., 2007). These injections are for instance estimated to be on the order of 10% in boreal regions (Hyer et al., 2007; Kahn et al., 2008), where they can be exceptionally exacerbated during violent pyro-convective events (Damoah et al., 2006; Fromm and Servranckx, 2003). The lack of vertical sensitivity of IASI to trace gas profiles does not allow for a precise estimate of the injection height and cannot confirm this: although useful information can be extracted from the CO vertical profiles in favourable situations (Turquety et al., 2009), this is unfortunately not the case for the observation discussed here, for which the retrieved CO profile does not reveal any fine structure on the vertical.

The species firmly (NH_3 , C_2H_4 , CH_3OH , HCOOH) or more tentatively (PAN, CH_3COOH) detected by IASI in the Siberia/Eastern Mongolia fire plumes are well known relatively short-lived biomass burning products (Andreae and Merlet, 2001), with lifetimes in the boundary layer ranging from a few hours (NH_3) to several days (CH_3OH), increasing for all species to several days/weeks at higher altitudes. These species have for instance been measured before by a variety of instruments, including for some ground- (Rinsland et al., 2005) or airborne-based infrared Fourier transform spectrometers (e.g. Yokelson et al., 1999, 2003; Worden et al., 1997). Their observations from space were, however, very sparse until recently. HCOOH (Rinsland et al., 2006), CH_3OH (Dufour et al., 2006, 2007) and C_2H_4 (Herbin et al., 2009) have now been extensively studied from the ACE-FTS solar occultation spectra in the upper troposphere (see also Rinsland et al., 2007). NH_3 and PAN have in addition been observed in a particular young biomass burning plume with the same instrument (Coheur et al., 2007) and by the MIPAS/Envisat limb emission spectrometer on larger scale (Burgess et al., 2006; Glatthor et al., 2007). More recently CH_3OH and NH_3 have been observed in a nadir mode by the Tropospheric Emission Spectrometer (TES) on a case basis (Beer et al., 2008). The results presented here thus provide the first observations of C_2H_4 , HCOOH , PAN and CH_3COOH for nadir sounding instruments. Furthermore, this is the first time that the simultaneous measurement of all above-mentioned species in fire plumes is made with such high spatial resolution, coverage and temporal sampling. As shown in the next section, this opens the way to support studies in relation to the chemistry processes in the fires plumes, which are currently mostly undertaken by confronting local ground based or airborne observations to sophisticated

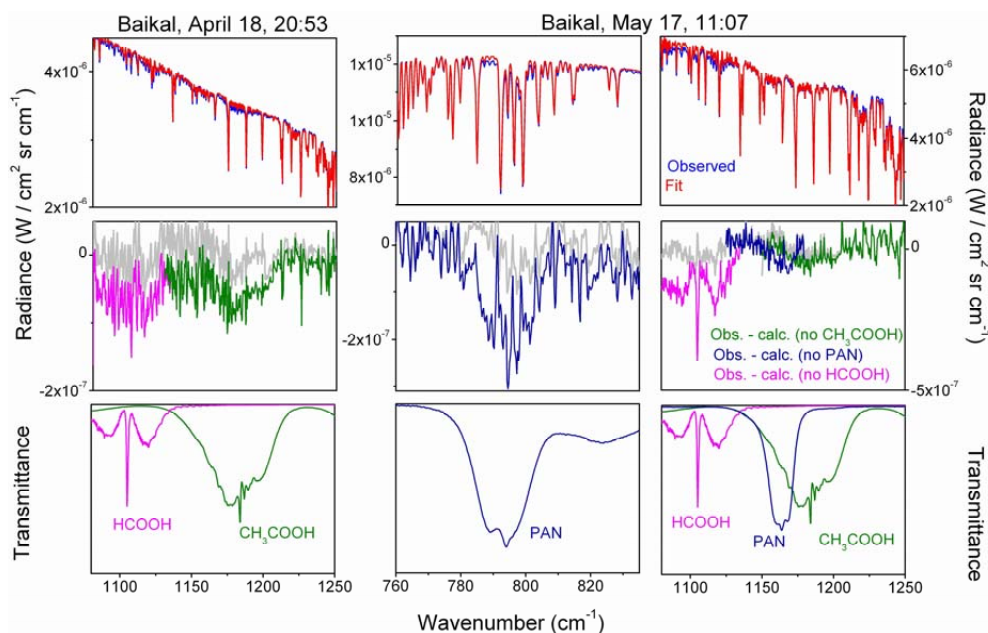


Fig. 4. Same as Fig. 3 (left panel) also shown for a spectrum measured on 17 May in Eastern Siberia (46.09° N, 118.99° E, 11:07 local time, middle and right vertical panels). The focus is given here to the region 1075–1250 cm⁻¹, with contributions from HCOOH ν_6 band, CH₃COOH and PAN (CO stretching modes), and to the region from 760 to 835 cm⁻¹ with contribution from PAN (NO₂ bending mode). In the middle panels, the light gray lines are the spectral residuals with all species accounted for in the retrieval, indicative of the best achievable RMS.

chemistry and transport models (Jost et al., 2003; Mason et al., 2001, 2006; Trentmann et al., 2003, 2005).

3.2 Emission and chemistry in the fire plumes

The large fires that occurred in Greece in the summer of 2007, which lasted for several days, are a good basis for performing a prospective study on the capabilities of IASI to track fire plumes from their emission source to regions downwind, as thoroughly discussed in a companion paper by Turquety et al. (2009), and also for capturing the chemical processes occurring in the first hours using the available information on the shorter lived species. Figure 5 (left panel) is similar to Fig. 3 in all respects, showing one typical IASI spectrum from the Greek fires on 25 August in the evening, when the emissions were largest, and the corresponding spectral fits in the atmospheric window. The NH₃, C₂H₄ and CH₃OH bands are clearly identified in the plume, transported here from Peloponnese to the Mediterranean Sea by North Eastern winds (Fig. 6). In contrast to the fires in South Siberia/Eastern Mongolia described previously, the spectral signatures of HCOOH, CH₃COOH and PAN are not detectable here, which could be explained by the larger amounts of VOCs emitted from boreal fires (Mason et al., 2006). For the particular spectrum shown in Fig. 5 we calculate total columns for NH₃, CH₃OH and C₂H₄ of, respectively $5.09 \pm 0.3 \cdot 10^{17}$, $6.78 \pm 2.0 \cdot 10^{16}$ and $1.22 \pm 1.6 \cdot 10^{17}$ molecules cm⁻², with volume mixing ratios

near the surface (essentially scaled versions of the prior profile due to the absence of vertical information) reaching 200 ppbv for NH₃, somewhat less for the two organic compounds. Elevated concentrations are also retrieved from IASI for CO, reaching $2.74 \cdot 10^{19}$ molecules cm⁻² with exceptional volume mixing ratios well above several ppmv in the boundary layer (Turquety et al., 2009). The range of column abundances retrieved from IASI can not directly be compared to other measurements inside biomass burning plumes, which sample the atmosphere only at low altitude.

The large plume of 25 August shown in Fig. 6 was found to be relatively localized at the time of the IASI first overpass in the morning (08:20 local time). It then extended over a much wider area in the evening (19:40 local time), reaching the African coasts on the 26th in the morning (between 08:00 and 09:40). This transport pattern is well seen in Fig. 7 on the basis of the NH₃ signal. It is fully consistent with the transport of CO and aerosols (Turquety et al., 2009). We find the highest concentrations in the different species in the plume from the 25th in the afternoon, showing that intense burning persisted during the entire daytime, lessening afterwards. The concentrations retrieved in the plume on the 26th in the morning have significantly decreased as a result of fast chemical processes but obviously also because of dilution in the ambient air. Assuming here that most of the plume measured on 25 August in the evening was emitted in a few hours, and simply integrating the concentrations

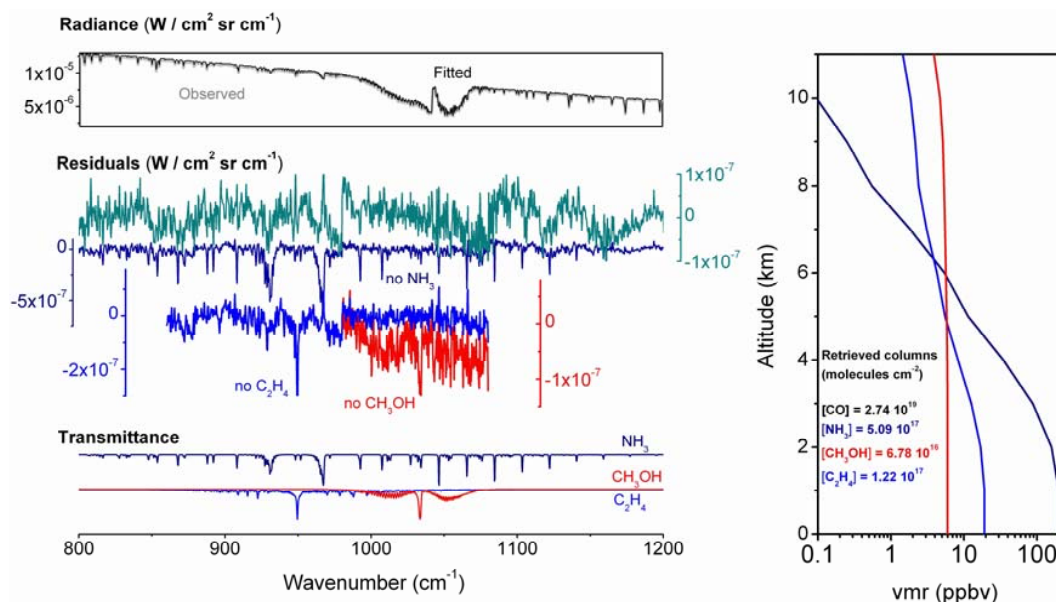


Fig. 5. Left: same as Fig. 3 for an observation of the fires from the Peloponnese on 25 August 2007 at 19:40 local time. Right: retrieved NH₃, CH₃OH and C₂H₄ profiles from that spectrum, in ppbv and on a logarithmic scale. The retrieved total columns are given inside the Figure, along with that of CO, obtained from FORLI-CO near-real-time processing (Turquety et al., 2009).

over the plume area, we estimate a total emitted mass for that particular day of $40 \cdot 10^3$, $6.5 \cdot 10^3$ and $7.0 \cdot 10^3$ Tons for NH₃, C₂H₄ and CH₃OH, respectively. Because of the already large uncertainties in the column retrieval for each individual pixels, these values are only indicative. Keeping this in mind, the NH₃ plume could represent a non negligible fraction (0.06%) of the yearly mean global emissions, and in particular of its biomass burning component (about 0.5%) (Galloway et al., 2004). For C₂H₄ and CH₃OH these would be smaller parts of the total emissions (0.025 and less than 0.01%, respectively; Folberth et al., 2006), which would still be considerable amounts in regard to the local and brief character of the event analyzed here.

Instead of looking into the concentrations of each species individually, the chemistry patterns in the fire plumes can best be highlighted by comparing the retrieved concentrations for the short-lived compounds to that of CO, which has a mean lifetime in the troposphere of several weeks. This procedure allows indeed removing the mixing component with ambient air, which occurs over time. This comparison is provided in Fig. 8 for the concentrations on the 25th in the evening and 26th in the morning. We find linear relations between CO and the other species for both days, with correlation coefficient above 0.9 for NH₃, 0.88 for C₂H₄ and around 0.8 for CH₃OH. Despite possible large errors on the determination of the column abundance for each species, which could in turn cause large errors on the enhancement ratios if only individual scenes were considered, the observed high degree of correlation among the species provides good confidence of the reproducibility on the column measurements

within a plume and as function of time. Hence, the value of the slopes $\partial[X]/\partial[\text{CO}]$, which give the enhancement ratio relative to CO, is more reliable than the column themselves. Close to the fire source, however, the linear regressions are not statistically representative because of the few measurements (see Fig. 7) and the calculated enhancement ratios are provided with larger errors. Values of 0.157, 0.039 and 0.009 for NH₃, C₂H₄ and CH₃OH are respectively calculated close to the source, which are extremely high for NH₃ and C₂H₄ as compared to those usually reported from aircraft measurements, even in fresh biomass burning plumes (e.g. Christian et al., 2007; Goode et al., 2000; Hobbs et al., 2003; Mauzerall et al., 1998; Yokelson et al., 2003). In fact such high values for the enhancement ratio of NH₃ relative to CO were only reported above very specific burning sources such as cattle dung and charcoal-making kilns (Christian et al., 2007). The temporal evolution of the enhancement ratio relative to CO over the 24 h period investigated here for the Greek fires is displayed in Fig. 9, on a logarithmic scale. There is a decrease for all species, which is consistent with the shorter lifetime of these compounds compared to that of CO. The values of the enhancement ratios for NH₃ and C₂H₄ drop very rapidly, by a factor of about seven, during the first twelve hours (Fig. 9) and slower afterwards. For CH₃OH the situation is similar but the rate of decrease during the first half of the period is much more reduced than for the other two species, which could be interpreted by less primary emission and possibly also secondary formation of that species in the plume (Holzinger et al., 2005).

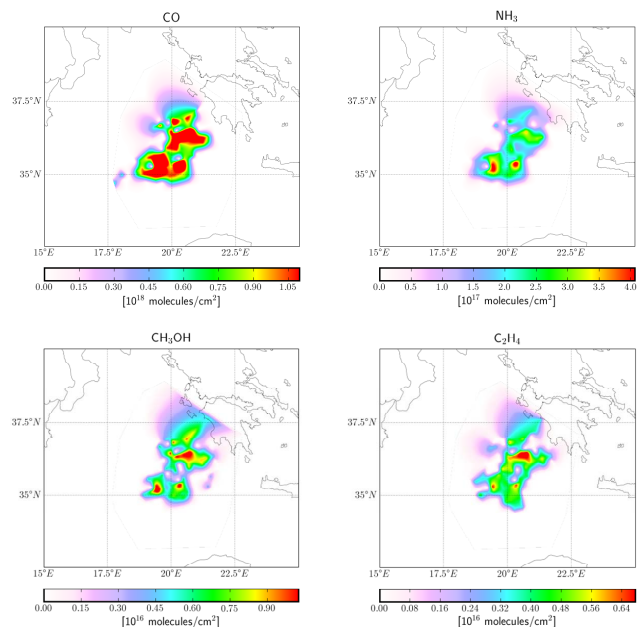


Fig. 6. Fire plume from the Peloponnese fires, measured on 25 August 2007 at 19:40 local time, shown separately from the upper left to the bottom right, for CO, NH₃, CH₃OH and C₂H₄ total columns, in molecules cm⁻². For the short-lived species only the measurements showing elevated BTD values were kept for the retrievals. The CO distribution was cut for the lower total columns (values smaller than 2.85×10^{18} cm⁻²) to reveal the enhanced concentrations in the plume from the background ones. Data are interpolated on a $0.125^\circ \times 0.125^\circ$ grid.

The analysis performed above for the plume of 25 to 26 August could unfortunately not be extended over a longer time period because the plume progressively disappeared and other emissions took place the following day in the same region. Similarly, such time-dependent study was difficult to perform for the South Siberia/East Mongolia fire events described above, which were not traceable in time, showing more of an intermittent character with the short-lived species being rapidly lost. As a basis of comparison, however, we find for the large plume observed on 17 May 2008, in Eastern Mongolia (see also Clerbaux et al., 2009), values of the $\partial[X]/\partial[CO]$ slopes of 0.013, 0.011 and 0.005 for NH₃, CH₃OH and C₂H₄, respectively, with, however, larger scatter around the linear regression (correlation coefficients between 0.53 and 0.73). These results suggest much lower ammonia and ethene emissions relative to CO than in the Greek fires.

4 Conclusions and perspectives

We have performed detailed analyses of thermal infrared radiance spectra measured from space in nadir geometry by the IASI/MetOp sounder above large forest fires. Two major events have been studied: the wildfires from August 2007 in

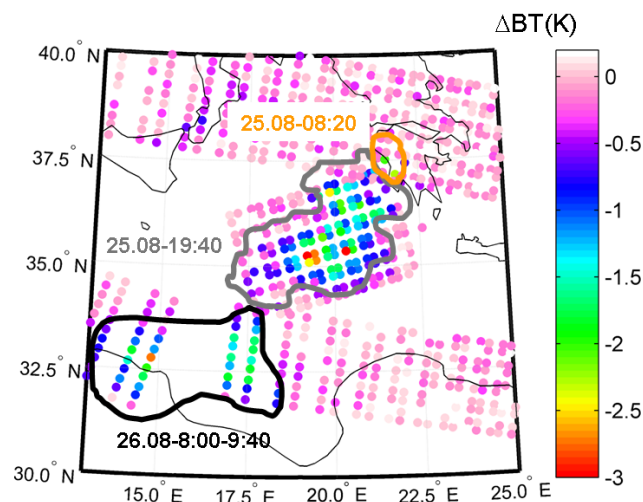


Fig. 7. IASI observation of the plume's transport from the Peloponnese fires during three successive orbits from 25 August 2007 in the morning to the next day, using NH₃ as tracer. Each point represents a IASI pixel, with the colour scale giving the BTD values associated to the NH₃ line at 867.75 cm^{-1} (negative values represent an absorption signal). Note that BTD values are given for a restricted number of measurements along the orbits; it is such as to give an estimate of the noise level outside the plume, while avoiding superposition of the values from different orbits. The orange, grey lines indicate the spatial extent of the plume for the three periods.

the Mediterranean basin, mainly originating from the Peloponnese peninsula, and those of early spring in Southern Siberia (Baikal Lake area) and Eastern Mongolia. In all these fire plumes, the presence of NH₃, C₂H₄ and CH₃OH has been firmly attested based on their spectral features in the atmospheric window. In the boreal fires, absorption bands of HCOOH and PAN have been observed, and the assignment of another broadband extinction feature to CH₃COOH has tentatively been made. For C₂H₄, HCOOH, and possibly PAN and CH₃COOH, these are the first reported observations from a nadir infrared sounder. Total columns have been retrieved for all species, with statistical errors generally in the range 5–30% (1σ), which are to be considered as a lower bound to the total error due to the limited vertical sensitivity and unknown plume height and in addition due to the possible influence of e.g. aerosols in these extreme environments. Taking advantage of the excellent spatial resolution, coverage and sampling of IASI, the horizontal extension of the plumes have been captured and their evolutions in space and time have been tracked. We have used as a case study the fire from the Peloponnese for three successive IASI orbits to provide insight onto the capabilities of IASI to contribute in understanding the chemical processes in the plume. Enhancement ratios of NH₃, CH₃OH and C₂H₄ relative to CO have been inferred by linear regression analysis and the values were found to be relatively high in comparison to literature data for a young biomass burning plume. The rapid

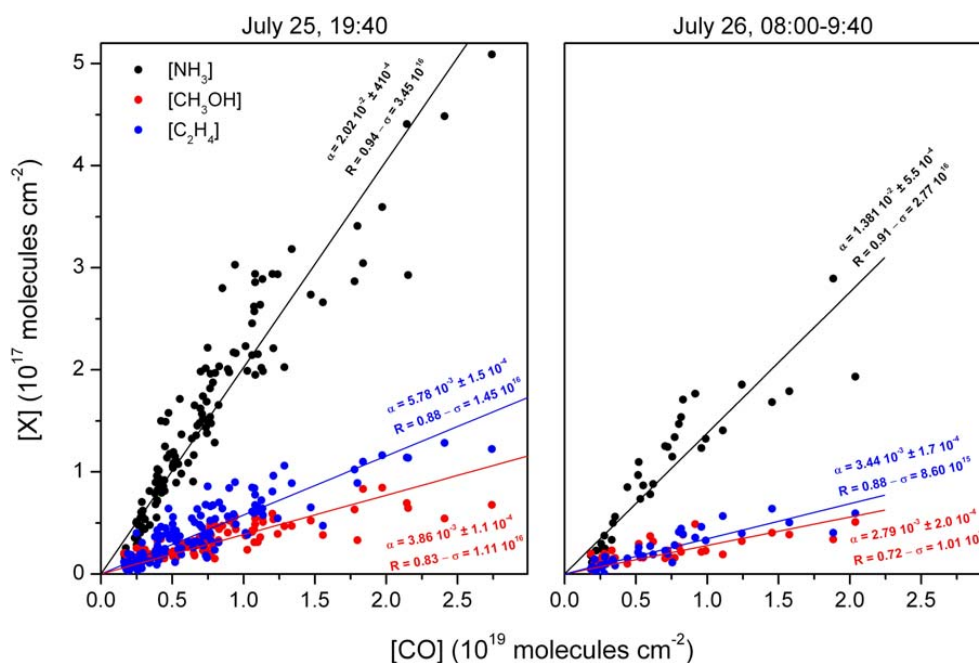


Fig. 8. Correlation between CO total columns and NH_3 , C_2H_4 and CH_3OH total columns retrieved in the fire plume on 25 August 2007 in the evening (left) and the 26 August 2007 in the morning (right). Linear regressions are plotted for each species vs. CO. The values for the resulting slopes and their errors are given in the figure, along with the correlation coefficient R and the standard deviation (1σ).

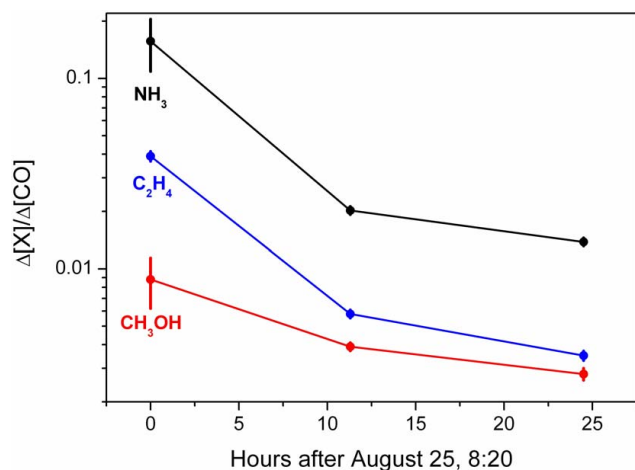


Fig. 9. Temporal evolution of the enhancement ratio of NH_3 , C_2H_4 , CH_3OH relative to CO, in logarithmic scale, during the 24 h after the plume's first observation on 25 August 2007, closest to the source region of the Peloponnese (see Fig. 7). The error bars are standard deviation (1σ) from the linear regressions relative to CO (see Fig. 8).

decrease of the enhancement ratios after half a day is explainable by the short lifetime of these three compounds relative to that of CO. A somewhat slower decrease of CH_3OH enhancement ratios as compared to that of NH_3 and C_2H_4 was found, possibly reflecting a secondary source of that species in the plume.

The analyses have suggested, using available information on the local thermal structure of the atmosphere, and assuming that the emitted plumes were largely confined to the lowest part of the atmosphere, that IASI was able to probe the chemical composition deep in the troposphere. If confirmed by more detailed forward and inverse simulations, this result would open extremely promising perspectives for identifying local to global sources of short-lived species and for monitoring air quality. This would obviously fill current observational gaps and improve our knowledge of both emission inventories and chemical reactions in the global atmospheric environment. As an example, a simple estimate of the NH_3 , C_2H_4 and CH_3OH mass emissions from the Peloponnese fires on 25 August 2007 was found to make up a non negligible contribution to the global emission budgets. Since the IASI launch in October 2006 several similar strong fires have been monitored and other emission sources have been revealed. It is therefore anticipated that the continuous monitoring of the lower atmosphere by IASI for the planned 14 years will ultimately help, along with global chemistry models, to better quantify the atmospheric budgets of a series of short-lived species. A first step in reaching this objective was recently achieved from IASI global observations of atmospheric ammonia (Clarisse et al., 2009a).

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